Magnetic linear dichroism effects in reflection spectroscopy: A case study at the Fe $M_{2,3}$ edge

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Abstract

Magneto-optical measurements are strongly dependent on the polarization of the radiation as well as the interaction geometry of the light with respect to the relative orientation and direction of the magnetization **M**. We performed magnetic circular dichroism (MCD) and magnetic linear dichroism (MLD) measurements around the Fe $M_{2,3}$ transition by measuring the difference between the reflected intensities of right and left circular polarized light (MCD) and the difference between the reflected intensities for opposite magnetization directions (MLD). From the angular variations of the MCD measurements we extracted the dielectric tensor $\varepsilon(\omega)$ which was then used as an input parameter to calculate the magneto-optical response of Fe in reflection MLD spectra. The experimental data around the $M_{2,3}$ edge show good agreement with model calculations.

Introduction

During the last few years magneto-optical measurements based upon the magneto-optical Kerr effect (MOKE) gained quite some popularity.[1-3] Magneto optical effects can be measured in a variety of different geometries using either circularly or linearly polarized radiation. Within the classical Fresnel-Maxwell framework, the magneto-optical effect can be described by the difference between the complex refractive indices for right- and left-circularly polarized light. The refractive index does affect the reflection amplitude and thus the intensities of the reflected light between the two opposite polarization or magnetization directions.

Magneto-optical effects are not restricted to circularly polarized radiation (MCD), but can also be measured with linearly polarized light under certain geometrical conditions. In order to create a difference signal in the reflectivity for the magnetic linear dichroism (MLD) measurement, the direction of the magnetization **M** needs to be changed. Even though the same dielectric tensor elements contribute to reflection MCD and MLD transitions, it is not straightforward to predict similarities in the spectral line shape, angular variations of the total intensity, or variations in the absolute integrated intensity of the MCD versus MLD spectra.

Due to the relative complexity involved in a first principals calculation of the energy dependence of the dielectric tensor $\varepsilon(\omega)$, there are no advanced model calculations discussing magneto-optical effects for reflection MCD or MLD from shallow core states. A slightly different approach based on a purely quantum mechanical model considering the different transition probabilities of polarized radiation from exchange and spin-orbit split atomic core states into free electron continuum states was first discussed by Thole and collaborators.[4] A recent paper by G. van der Laan[5], which extends the earlier model calculations, seems to reasonably describe the main features seen in MLD-photoemission experiments.[6,7] The results of the one-electron calculations[4-7] indicate significant differences between the photoemission line shapes of angular resolved MCD and MLD experiments. Unfortunately, these theories are not directly applicable to reflection based magneto-optical measurements performed as a function of photon energy rather than at a constant photon energy as in photoemission.

In previous experiments we measured the MCD effect around the $M_{2,3}$ edges of ferromagnetic films and developed a method to extract the dielectric tensor elements from the angular variation of the MCD spectra.[8] These experimentally determined off-diagonal tensor elements ε_{xy} of Fe were used in the present study to test calculations based on the classical Fresnel-Maxwell formalism for magneto-optical effects involving linearly polarized radiation. We report experiments confirming our MLD model calculations predicting transversal MLD-effects which are considerably stronger than those measured with circularly polarized light.

Experimental Procedures

Due to the symmetry relation of the magneto-optical transitions, experiments can be performed by either keeping the direction of magnetization **M** fixed while changing the light helicity between right and left circular, or by alternating the direction of magnetization **M** while holding the helicity of light constant. In the MCD experiments we preferred to keep the orientation of the magnetization fixed while modulating the helicity of the light, whereas for the MLD experiments the magnetization direction is the only parameter which can be modulated to generate a difference signal in the reflection intensities.

For the MCD experiments we used a quadruple reflection polarizer to convert monochromatic linearly polarized synchrotron radiation into circular light.[9] By selecting the proper angles of reflection, it is possible to maintain a total phase shift of Δ_{sp}=± 90° between the two perpendicular reflection coefficients r_s and r_p that converts the linearly polarized light beam (P_{lin} = 0.99) into a right or left circular polarized light beam (P_{circ} = 0.98). Further information regarding beamline characteristics such as photon flux and energy resolution and design details of the quadruple reflection polarizer and a performance evaluation can be found in Refs. 9–11. The magnetic linear dichroism experiments were also performed with the quadruple reflector, but in a geometry where the four consecutive reflecting surfaces are in the horizontal plane (the plane of the storage ring orbit). Since in this geometry only the r_s component is transmitted, the presence of the quadruple reflection polarizer further enhances the already high degree of polarization of the horizontally linear polarized light beam of monochromatized synchrotron radiation.

The experiments were performed at the University of Wisconsin's Synchrotron Radiation Center using the Amoco 6m toroidal grating monochromator beamline.[11] The quadruple reflection polarizer was installed at the exit mirror box of the beamline to allow for variable light polarization. The light intensity reflected off the sample's surface is measured with a Si-photodiode. The sample and diode are mounted on precision rotary manipulators allowing to separately adjust both angles of incidence. Intensity scans are taken at a fixed angle of incidence θ while scanning the photon energy. To account for beam fluctuations and natural beam decay during the course of a storage ring fill, the Si-diode current is normalized with a signal obtained from a Ni-mesh which is located between the beamline exit slit and refocusing mirror box.

Thin Fe films of ~2000 Å thickness were deposited at room temperature under ultra-high vacuum conditions on mirror-polished (0.05 µm grit) stainless-steel substrates. The films were permanently magnetized in the plane of the sample by rare earth magnets which produce a magnetic field of ~0.3 T at the sample surface. For the MLD measurements the sample/magnet assembly was rotated around the x-axis to change the magnetization direction along the z-axis.The interaction geometry is shown schematically in Figure 1.

Brief outline of the MLD formalism

The basic features of the different magneto-optical effects, which are the response of the reflected or absorbed light to angular and energetic variations, can be described within the classical Maxwell theory. Using this approach has the benefit of working within a set of simple equations which directly relate dielectric optical properties to measurable quantities such as the absorbed or reflected intensity.[12] For the current study, where we were simply interested in understanding the differences and relative merits of magneto-optical effects measured under various experimental geometries, the classical approach seems to be convenient and easy to follow.

The price one pays for this apparent comfort is that the extracted information is now hidden in the frequency dependencies of the dielectric tensor elements from which it is not straight forward to directly extract microscopic material properties such as the magnetic moment μ and its decomposition into the respective spin $<S_z>$ and orbital $<L_z>$ components via the so-called sum rules. Nevertheless, we have investigated sum rule with regard to our classical approach and will report our the findings elsewhere.[13]

For the reflected light MLD experiments the polarization of the light wave was in the xy-plane of the sample which is perpendicular to the direction of magnetization as shown in Fig. 1. In turn, the magnetization **M** is parallel to the sample surface along the z-direction. Magnetizing the sample **M** causes anisotropy and changes the dielectric properties of the thin film from scalar to tensor which has the general form:

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & 0 \\ -\varepsilon_{xy} & \varepsilon_{xx} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{pmatrix}$$

For such a system the refractive index n is to the first order given by the diagonal element ε_{xx} of the dielectric tensor

$$\hat{n} = \sqrt{\varepsilon_{xx}}$$

and the complex refraction angle χ is given by

$$\sin(\chi) = \sin(\theta) / \hat{n}$$

The boundary conditions at the surface require that the tangential component of the electric field vector **E** and the magnetic field vector **H** are continuous. One obtains a set of two equations which allow the calculation of the complex reflection amplitude R_p.

$$R_p = \frac{n\cos(\theta) - \cos(\chi)}{n\cos(\theta) + \cos(\chi)} A_p - 2\frac{\hat{\varepsilon}_{xy}\cos(\theta)\sin(\chi)}{n(n\cos(\theta) + \cos(\chi))^2} A_p$$

where n and ε_{xy} are the previously defined refractive index and the off-diagonal dielectric tensor elements.

The reflected intensities $I^{\pm}$, where $\pm$ indicates the two opposite directions of magnetization **M** along the z-axis, is given by

$$I^{+} = |R_p|^2 I_0$$

The strength of the MLD-effect is now defined as the difference of the reflected light intensities $I^{\pm}$ between two opposite magnetization directions M^{+} and M^{-}, normalized to the average of the total reflected intensity,

$$MLD - effect = \frac{(I^{+} - I^{-})}{1/2(I^{+} + I^{-})}$$

The factor of 1/2 is commonly used in dichroism experiments to normalize to unit intensity.

Results and Discussion

In a previous paper we reported the reflection MCD effect of ferromagnetic Fe films around the Fe $M_{2,3}$ edge.[8] Using the Fresnel-Maxwell equations we were able to fit the angular dependence of the MCD spectra by means of an energy dependent dielectric tensor $\varepsilon(\omega)$ where the diagonal elements $\varepsilon_{xx}(\omega)$ represent the non-magnetic optical properties of the Fe film. We chose to use literature values of the dielectric constants *n* and *k*[14] from which we computed $\varepsilon_{xx}(\omega)$ rather than determining them as additional free parameters in the MCD fits.

Figure 2 shows the real and imaginary part of the off-diagonal elements $\varepsilon_{xy}(\omega)$ that result from the fitting routine employed in Ref. 8. The error bars result from the uncertainties of the least square fit of $\varepsilon_{xy}(\omega)$ and the assumption that *n* and *k* have a constant error of ±5%. This error in *n* and *k* was assumed in order to study the propagation of errors introduced by the diagonal elements in the least squares fitting of the off-diagonal elements. We found that errors of this size do not change the characteristic features of $\varepsilon_{xy}(\omega)$, thus indicating that our analysis determines $\varepsilon_{xy}(\omega)$ as a unique material characteristic rather than as a parameter that is simply constructed from $\varepsilon_{xx}(\omega)$.

The spectral shape of the real and imaginary part have similarities with what is referred to as a “diamagnetic” or type 1 lineshape: the real part $\varepsilon^1{}_{xy}(\omega)$ peaks around the transition energy with negative underswings prior to and after the transition, while the complex part $\varepsilon^2{}_{xy}(\omega)$ appears like the derivative of an absorption peak. The “diamagnetic” line shape is the result of a single initial state transition with equal but opposite oscillator strength for right and left circular polarized light into split final states.

[14,15] Since the energy dependence of the tensor elements directly resembles the matrix elements of optical transitions into spin orbit split final states of a real multi electron system, it is not to be expected that the ε_{xy} Fe-data exactly follow the shape of either the type 1 or a so called type 2 or “paramagnetic” line shape. The observed line shape deviations from these simple models are of course a result of the details of the spin polarized conduction band structure of Fe.

In the following we will use the dielectric tensor elements shown in Fig. 2, which were determined from the longitudinal MCD experiments, as input parameters for the MLD model equations in order to test the predictions using experimental MLD measurements. The spectra I^+ and I^- are separately measured as a function of photon energy while keeping the angle of incidence θ constant. A typical spectrum displaying the variation of the reflected intensities as well as the resulting MLD-effect at an incidence angle of θ=45° is shown in Figure 3. Similar to typical MCD line shapes (*c.f.* Fig. 2 in Ref. 8), the MLD-effect also shows the derivative type spectral shape centered around the $M_{2,3}$ edge.

There is, however, a noticeable difference: the MLD-effect is considerably larger than the longitudinal MCD-effect which is calculated using the same definition as the MLD effect. (A complementary comparison between the longitudinal and polar MCD effects will be reported elsewhere.[17]) This finding, which seems at first unreasonable since the same basic tensor elements contribute to the optical transitions, has its origin in the different behavior between the angular variation of the numerator and denominator used to compute the MLD-effect. It turns out that the reflection intensities I^+ and I^- approach each other and cross-over in the reflection minimum region around θ=50-45° causing the sum of the intensities (denominator) to drop faster than the intensity difference (numerator).

Compared to the minor differences between the MLD and MCD-line shapes the “resonant behavior” of the MLD effect around θ=45°seems to be the more prominent fingerprint. To quantify our observations we calculated the integral of the absolute magnitude of the MLD-effect over the range of photon energy (from hν = 46 eV to hν = 62 eV for the $M_{2,3}$ absorption edge) where the effect is non-zero. Figure 4 shows a comparison of the integrated experimental curves with the theoretical predictions based on the MLD model calculation as a function of the angle of incidence θ. For comparison we also show the integrated absolute MCD intensities. The MCD experiment was performed in the longitudinal geometry, where the magnetization is also in the plane of the sample but oriented parallel to the direction of light propagation. The dashed line which closely follows the experimental lonitudinal MCD data points is the calculated angular dependance based upon the tensor elements shown in Fig. 2. Both sets of experimental data clearly follow the predicted angular variations with distinct maxima around θ~45° for MLD and ~60° for the longitudinal MCD data.

However, it should be noted that the absolute values of the experimentally determined MLD data are somewhat bigger than the theoretical prediction and are scaled by a factor of 0.85 to match the theory. The reason for this discrepancy could be related to differences in the optical properties of the sample surface caused by varying growth and vacuum conditions during the two types of experiments. The MLD and MCD experiments where performed in different vacuum chambers utilizing a MBE effusion cell and an e-beam evaporator. Even though we were generally concerned about sample contamination and frequently redeposited Fe films to minimize surface contamination, it can not be ruled out that both experiments had Fe films of slightly different overlayer properties. In both experiments we noticed the adverse effects of sample contamination either by depositing Fe films under slightly enhanced background pressures or by degradation of the surface cleanliness due to surface contamination/oxidation effects.

Conclusions

Using previously determined off-diagonal dielectric tensor elements $\varepsilon_{xy}(\omega)$ from reflection MCD experiments around the $M_{2,3}$ region of Fe we have verified the validity of model calculations based on the classical dielctric theory. We measured the magnetic linear dichroism effect (MLD) over a large angular range and compared the results with the theoretical predictions. The good overall agreement between MLD theory and experiment can be interpreted as a self-consistency check of the model equations describing the magneto-optical effects in various interacting geometries.

Our data also show the potential of MLD spectroscopy which provides the same basic dielectric tensor information as one might extract from MCD experiments. Considering the efforts to generate and maintain constant properties of right and/or left circularly polarized light compared to the ease in utilizing the readily available strong linearly polarized light component of the synchrotron radiation fan, MLD measurements should be considered the experiment of choice in reflection type magneto-optical studies.

Acknowledgments

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Figure Captions

Figure 1: Definition of the interaction geometry for transversal MLD measurements.

Figure 2: Experimentally determined energy dependence of the real and complex dielectric constants $\varepsilon_{xy}(\omega)$ Fe around the $M_{2,3}$ transition region of Fe.

Figure 3: (a) Energy dependence of the transversal MLD intensity spectra $I^{\pm}$ and (b) the resulting normalized MLD-effect of Fe measured at an angle of incidence θ=45°.

Figure 4: Comparison between the calculated angular variation of the integral MLD and MCD-effect with experimentally determined integral values.

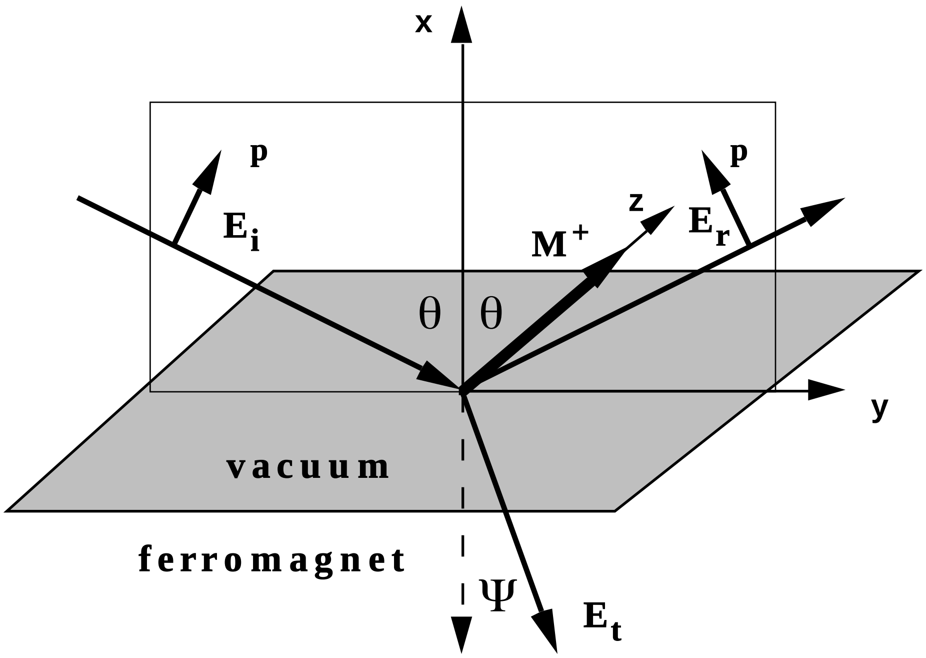
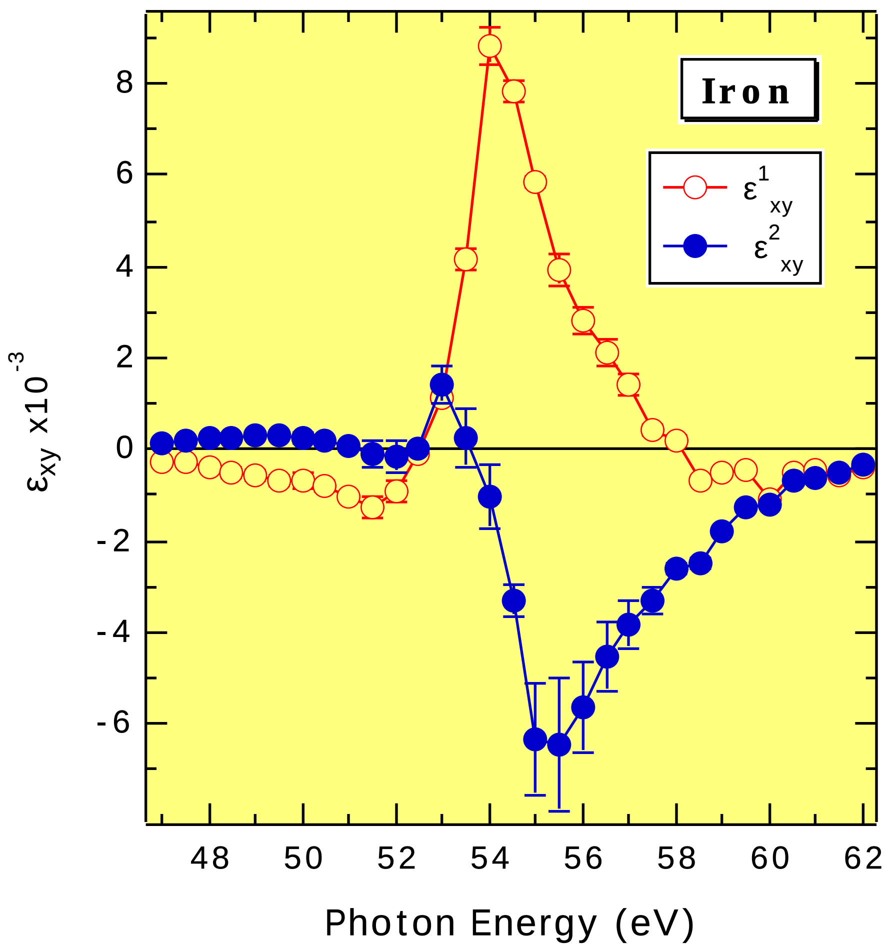
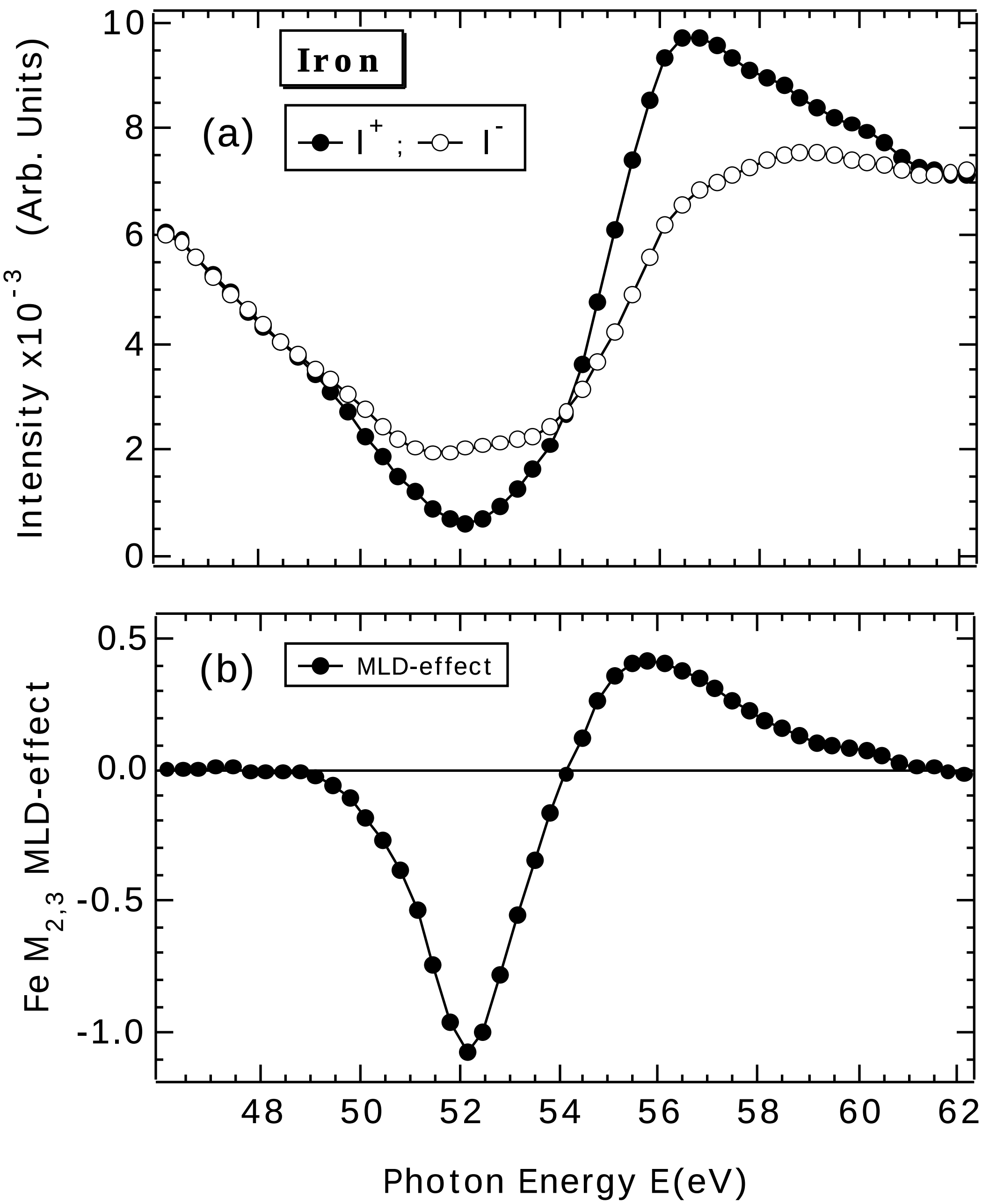
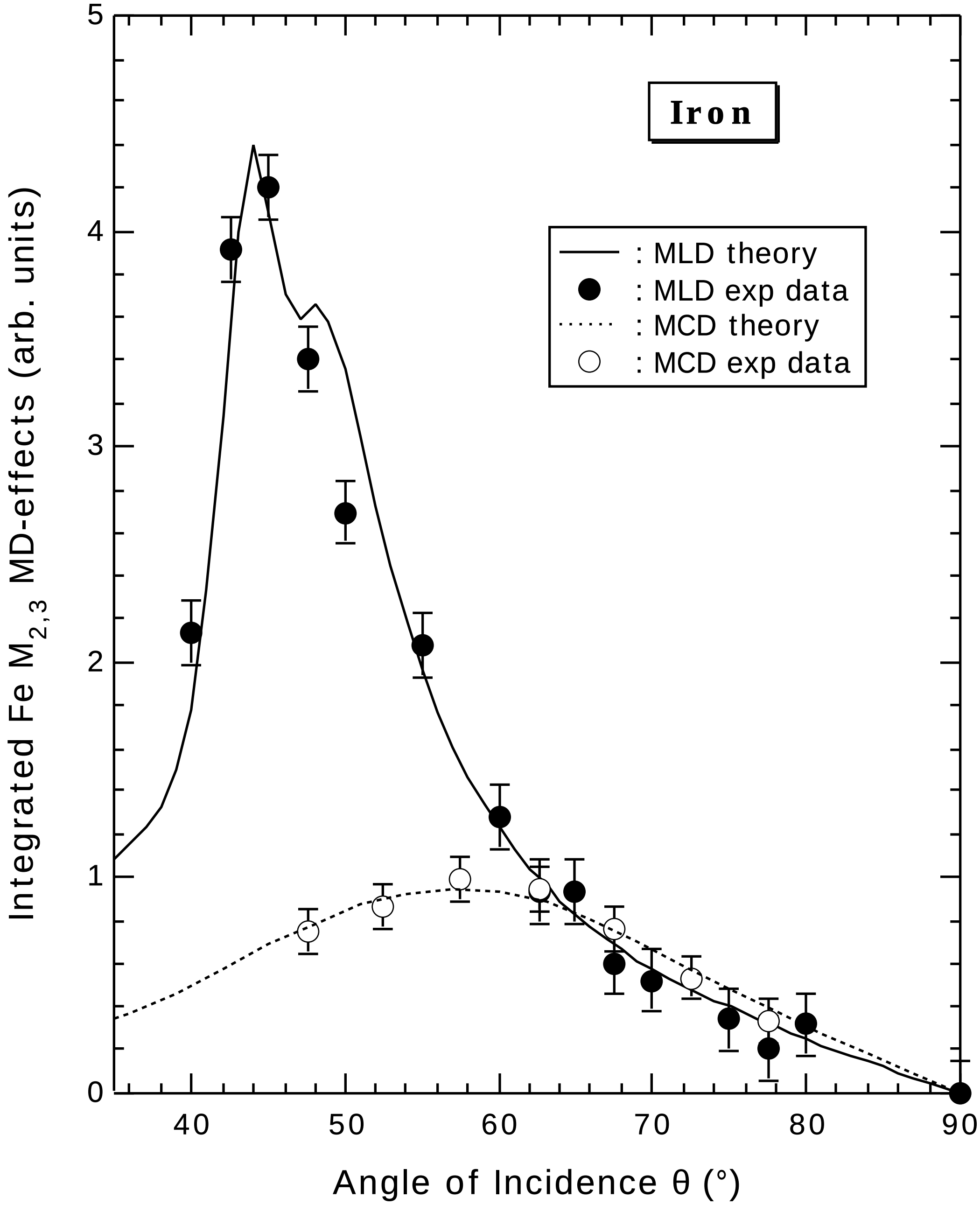


Figure 1


Iron
ε^1_{xy}
ε^2_{xy}
ε_{xy} x10^{-3}
8
6
4
2
0
-2
-4
-6
48
50
52
54
56
58
60
62
Photon Energy (eV)



Iron
(a)
I+ ; I-
Intensity x10-3 (Arb. Units)
0
2
4
6
8
10
(b)
MLD-effect
Fe M2,3 MLD-effect
0.5
0.0
-0.5
-1.0
48
50
52
54
56
58
60
62
Photon Energy E(eV)


Iron
: MLD theory
: MLD exp data
: MCD theory
: MCD exp data
Integrated Fe $M_{2,3}$ MD-effects (arb. units)
Angle of Incidence θ (°)
0
1
2
3
4
5
40
50
60
70
80
90